Bending admissible continuum approximation for mono-layered 2D materials

Xingjie Helen Li

University of North Carolina at Charlotte

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Collaborators: Julian Braun (University of Warwick, UK)
Derek Olson (Rensselaer Polytechnic Institute)

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Outline

- Motivation: non-flat configuration

- Continuum model: von-Kármán for mono-layered system
  - assumptions on the empirical atomistic energy
  - scaling rules of the displacement fields
  - appropriate norm that reflects the observed stability

- Stability and error analysis
  - linear stability analysis
  - force consistency analysis
  - displacement fields error estimate

- Ongoing work

- Summary
Experimental observations

Graphene is locally two-dimensional but not flat. Nanoscale ripples appear in suspended samples and rolling up often occurs when boundaries are not fixed. 1 2

(a) Ripple 3

(b) Rolling up 4

Need an appropriate continuum model for multiscale modelings.

3 Deng and Berry, Materials Today, Volume 19 (4), (2016)
A few existing continuum models

- Based on Cauchy–Born rule
  - Exponential Cauchy–Born rule; 5
  - High order Cauchy–Born rule. 6 7

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5 Arroyo and Belytschko, 40, 455-469, Meccanica, (2005)
7 Cazeaux and Luskin, M2AN, 52, 729-749, (2018)
9 Yang and Tewary, PRB 77, 245442, (2008)
10 Xiaojie Wu and Xiantao Li, manuscript
A few existing continuum models

- **Based on Cauchy–Born rule**
  - Exponential Cauchy–Born rule; ⁵
  - High order Cauchy–Born rule. ⁶ ⁷

- **Based on the harmonic approach**
  - Iterative method by using force corrections; ⁸
  - Discrete lattice Green function and obtain the linear Kirchhoff plate theory for the far-field considering a point defect/ point force loading. ⁹ ¹⁰

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- **Based on nonlinear plate model**
  - For example, using the *von-Kármán plate* theory. ¹¹ ¹²

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Some of them do not target for modeling ripples.

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Literatures related to the von-Kármán theory

- **Applicational point of view:** The von-Kármán has been used for modeling and computational simulations of ripples and wrinkles, see for example, Kumar et. al. (2015)\textsuperscript{13} and Gao et. al. (2019).\textsuperscript{14}

\textsuperscript{13}Kumar et. al. Scientific Reports, 5:10872, (2015)
\textsuperscript{14}Gao et. al. Handbook of Graphene: 2, 1-44, (2019)
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- **Multilayered graphene system:** Braun et. al. rigorously derived the von-Kármán model via analyzing scaling of energy in terms of the lattice constant \(\varepsilon\) and the layer distance \(h\). \(^{15}\)

- **Continuum plate theories:** Friesecke et. al. rigorously studied different plate models by scaling of the elastic energy per unit volume in terms of the thickness \(h\) of plates \(^{16}\)
  - \(f^{\text{ext}}\) and total energy \(\sim h^2\): Kirchhoff plate
  - \(f^{\text{ext}}\) and total energy \(\sim h^4\): von-Kármán plate
  - general case with external force \(\sim h^\alpha\) with \(\alpha > 0\), summarized in Table 1 of Friesecke et. al. (2006)

\(^{13}\) Kumar et. al. Scientific Reports, 5:10872, (2015)
Goal

1. Rigorously derive the continuum von-Kármán model which captures the governing out-of-plane rippling type deformations in a mono-layered system.

2. Prove rigorous stability and error analysis.
Consider a simple lattice with reference configuration

\[ \mathcal{L} := \varepsilon F \mathbb{Z}^d, \quad \text{for some } F \in \mathbb{R}^{d \times d}, \quad \det(F) = 1, \quad d \in \{1, 2\}, \]

where \( \varepsilon \) is the lattice constant. It is deformed into \( \mathbf{y} : \mathcal{L} \to \mathbb{R}^{d+1} \).

For simplicity, consider a 1D periodic chain of \( N \in \mathbb{N} \) atoms. Assume \( \varepsilon N = 1 \), thus the reference atomic chain is \( \mathcal{L} = (0, 1] \).

Denote the physical displacement field by \( \mathbf{u} : \mathcal{L} \to \mathbb{R}^2 \) where

\[
\mathbf{u}(\varepsilon i) := \mathbf{y}(\varepsilon i) - \begin{pmatrix} \varepsilon i \\ 0 \end{pmatrix} = \begin{pmatrix} u_1(\varepsilon i) \\ u_2(\varepsilon i) \end{pmatrix} = \begin{pmatrix} u_{i,1} \\ u_{i,2} \end{pmatrix}.
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\[ u(\varepsilon i) := y(\varepsilon i) - \begin{pmatrix} \varepsilon i \\ 0 \end{pmatrix} = \begin{pmatrix} u_1(\varepsilon i) \\ u_2(\varepsilon i) \end{pmatrix} = \begin{pmatrix} u_{i,1} \\ u_{i,2} \end{pmatrix}. \]

For bulk materials, \( u_1 \) and \( u_2 \) are scaled in the same way with respect to \( \varepsilon \).

Challenge: For 2D materials, need to identify different scaling rules of \( u_1 \) and \( u_2 \) wrt \( \varepsilon \).
Atomistic energy

The total atomistic energy for 1D chain is

\[ E^a(y) = E_{pp}^a(y) + E_{ap}^a(y) = \varepsilon \sum_{i=1}^{N} \left[ V \left( \frac{|y_{i+1} - y_i|}{\varepsilon} \right) + f(\theta_i) \right]. \]

\( E_{pp}^a \) denotes the pairwise potential and \( E_{ap}^a \) denotes the angular potential.

We assume the following properties:

- \( V(s) \) behaves like a quadratic function, it reaches local minimum at \( s = 1 \) with \( V(1) = V'(1) = 0 \) and \( V''(1) > 0 \);
- \( \theta_i := \theta(i) \) measures the angle between vector \( \{y_i - y_{i-1}\} \) and vector \( \{y_{i+1} - y_i\} \) and \( 0 < \theta_i \ll 1 \);
- \( f(\theta) \) denotes the angular potential that also behaves like a quadratic function with local minimum at \( \theta = 0 \), and \( f(0) = f'(0) = 0 \), \( f''(0) > 0 \), \( f(\theta) \approx \theta^2 \) when \( 0 < \theta \ll 1 \).
Scaling of displacement field

- Total energy is scaled as $\sim \varepsilon^4$ for the von-Kármán plate:
  $$E^a(y) \leq C\varepsilon^4.$$ 

We then proved that the deformed configuration $y$ is close to a rigid motion: $x \rightarrow R(x + c)$, where $c$ denotes a translation and $R$ denotes a rotation.
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- Hence, the displacement field $u$ must be scaled as

$$u = \begin{pmatrix} u_1 \\ u_2 \end{pmatrix} = \begin{pmatrix} \varepsilon^2 \tilde{u}_1 \\ \varepsilon \tilde{u}_2 \end{pmatrix},$$

where $\tilde{u}_1, \tilde{u}_2$ are scalar functions and are independent of $\varepsilon$. 

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where $\tilde{u}_1$, $\tilde{u}_2$ are scalar functions and are independent of $\varepsilon$.

- Hence $\frac{1}{\varepsilon^4} E^a$ is $\Gamma$–convergent to the von-Kármán energy as $\varepsilon \to 0$

$$E^c(\hat{u}) = E^c(\hat{u}_1(x), \hat{u}_2(x)) = \int_0^1 \left[ \frac{1}{2} V''(1) \left( \hat{u}_1'(x) + \frac{1}{2}(\hat{u}_2'(x))^2 \right)^2 \\
\quad + \frac{1}{2} f''(0)(\hat{u}_2''(x))^2 \right] dx.$$  

$\hat{u}_1(x)$ and $\hat{u}_2(x)$ are smooth interpolation of the scaled displacements $\tilde{u}_1$ and $\tilde{u}_2$ from $L \to \mathbb{R}$.  

9
Appropriate norm for the observed stability

Define the first order differentiation operator

\[ D_1^\epsilon v_\ell = \frac{v_{\ell+1} - v_\ell}{\epsilon}, \]

and the second order differentiation operator

\[ D_{1(2)}^\epsilon v_\ell = \frac{D_1^\epsilon v_\ell - D_1^\epsilon v_{\ell-1}}{\epsilon} = \frac{v_{\ell+1} - 2v_\ell + v_{\ell-1}}{\epsilon^2}. \]

For a physical displacement field \( u \), the semi-norm is defined as

\[ \|u\|_*^2 := \|D_1^\epsilon u_1\|_{\ell_2^\epsilon}^2 + \epsilon^2 \|D_{1(2)}^\epsilon u_2\|_{\ell_2^\epsilon}^2, \]

where for any scalar function \( v \)

\[ \|v\|_{\ell_2^\epsilon}^2 = \epsilon \sum_{i=1}^N |v_i|^2. \]

For a continuum scaled displacement field \( \hat{u} \), the semi-norm is defined as

\[ \|\hat{u}\|_{*c}^2 := \int_0^1 \left[ (\hat{u}_1')^2 + (\hat{u}_2'')^2 \right] dx. \]
Linear stability

- The atomistic model is linear stable wrt to the $\| \cdot \|_*$:
  \[
  \langle \delta^2 E^a(0)v, v \rangle \geq \gamma^a \|v\|^2_*.
  \]

- The continuum model is linear stable wrt to the $\| \cdot \|_{*c}$:
  \[
  \langle \delta^2 E^c(0)\hat{v}, \hat{v} \rangle \geq (1 + \varepsilon^2)\gamma^a \|\hat{v}\|^2_{*c}.
  \]

- Consequently, both atomistic and continuum models are linear stable and with similar stability constants.
Approximation error

Approximation error in terms of energy difference.

Applying Taylor expansion to the atomic site energy of site $i$:

Pair interaction:

$$V(u_i) = V \left( \sqrt{(1 + D_1^\epsilon u_{i,1})^2 + (D_1^\epsilon u_{i,2})^2} \right)$$

$$\approx \frac{1}{2} V''(1) \left( D_1^\epsilon u_{i,1} + \frac{1}{2} (D_1^\epsilon u_{i,2})^2 \right)^2 + O(\epsilon^6)$$

$$= \epsilon^4 \frac{1}{2} V''(1) \left( D_1^\epsilon \tilde{u}_{i,1} + \frac{1}{2} (D_1^\epsilon \tilde{u}_{i,2})^2 \right)^2 + O(\epsilon^6).$$

Angular interaction:

$$f(\theta_i) \approx f''(0) \theta_i^2 \approx \epsilon^2 f''(0) \left( D_1^{(2),\epsilon} u_{i,2} \right)^2 + O(\epsilon^6)$$

$$= \epsilon^4 f''(0) \left( D_1^{(2),\epsilon} \tilde{u}_{i,2} \right)^2 + O(\epsilon^6).$$

Hence, the continuum total energy is of second order consistent comparing to the scaled atomic total energy.
Approximation error

**Approximation error in terms of energy difference.**

- Applying Taylor expansion to the atomic site energy of site $i$:
  
  **Pair interaction:**
  
  $$V(u_i) = V \left( \sqrt{1 + D_1^\varepsilon u_{i,1}^2 + (D_1^\varepsilon u_{i,2}^2)} \right)$$
  
  $$\approx \frac{1}{2} V''(1) \left( D_1^\varepsilon u_{i,1} + \frac{1}{2} (D_1^\varepsilon u_{i,2}^2) \right)^2 + O(\varepsilon^6)$$
  
  $$= \varepsilon^4 \frac{1}{2} V''(1) \left( D_1^\varepsilon \tilde{u}_{i,1} + \frac{1}{2} (D_1^\varepsilon \tilde{u}_{i,2}^2) \right)^2 + O(\varepsilon^6).$$

  **Angular interaction:**
  
  $$f(\theta_i) \approx f''(0) \theta_i^2 \approx \varepsilon^2 f''(0) \left( D_1^{(2)} \varepsilon u_{i,2}^2 \right) + O(\varepsilon^6)$$
  
  $$= \varepsilon^4 f''(0) \left( D_1^{(2)} \varepsilon \tilde{u}_{i,2}^2 \right)^2 + O(\varepsilon^6).$$

- Hence, **the continuum total energy is of second order consistent comparing to the scaled atomic total energy.**
Approximation error

**Approximation error in terms of force difference.**

- Compute $F^a_i(u)$ and get

$$F^a_i(u) = -\frac{1}{\varepsilon} \frac{\partial E^a}{\partial u_i}$$

$$= V''(1) \begin{pmatrix} \varepsilon^2 (\tilde{u}''_{i,1} + \tilde{u}'_{i,2} \tilde{u}''_{i,2} + O(\varepsilon)) \\ \varepsilon^3 (\tilde{u}_{i,2} \tilde{u}'_{i,1} + \tilde{u}'_{i,2} \tilde{u}''_{i,1} + \frac{3}{2} \tilde{u}''_{i,2} (\tilde{u}'_{i,2})^2 + O(\varepsilon)) \end{pmatrix}$$

$$+ f''(0) \begin{pmatrix} 0 + O(\varepsilon^3) \\ \varepsilon^3 (\tilde{u}^{(4)}_{i,2} + O(\varepsilon)) \end{pmatrix}$$

$$= \begin{pmatrix} \varepsilon^2 (F^c_{i,1}(\tilde{u}) + O(\varepsilon)) \\ \varepsilon^3 (F^c_{i,2}(\tilde{u}) + O(\varepsilon)) \end{pmatrix}.$$

- Hence, the pointwise error between the scaled $F^a_i$ and the $F^c_i$ is of first order.

- As a result, the scaled continuum displacement field has first order modeling error wrt atomistic $\| \cdot \|_\ast$. 
Extension to 2D

- Suppose $y : \varepsilon \mathbb{Z}^2 \to \mathbb{R}^3$ and $E^a \leq C \varepsilon^4$ with

$$E^a(y) = \varepsilon^2 \sum_i \left( V(D_1^\varepsilon y_i) + f(\theta_i) + g(\omega_i) \right),$$

where $V$ denotes the pair potential, $f$ denotes the bond angle $\theta$ potential and $g$ denotes torsion angle $\omega$ (angle between two triangles) potential.

- Assume a similar scaling rule of displacement field

$$u = \begin{pmatrix} u_1 \\ u_2 \\ u_3 \end{pmatrix} = \begin{pmatrix} \varepsilon^2 \tilde{u}_1 \\ \varepsilon^2 u_2 \\ \varepsilon u_3 \end{pmatrix}.$$

- So $\frac{1}{\varepsilon^4} E^a$ is $\Gamma$-convergent to the von-Kármán model

$$E^c(\tilde{u}) = \int \mathbb{C} \left( \text{sym}(\nabla \tilde{u}_{1,2}) + \frac{1}{2} \nabla \tilde{u}_3 \otimes \nabla u_3 \right)$$

$$\cdot \left( \text{sym}(\nabla \tilde{u}_{1,2}) + \frac{1}{2} \nabla \tilde{u}_3 \otimes \nabla \tilde{u}_3 \right) \, dx_1 \, dx_2$$

$$+ \int \frac{k}{2} |\nabla^2 \tilde{u}_3|^2 \, dx_1 \, dx_2.$$
Extension to 2D

Four-body interactions must be included; and Tersoff and Brenner type interatomic potentials satisfy the assumptions. Analysis is similar to the 1D case.

- The continuum total energy is second order consistent comparing to the scaled atomic total energy.
- The continuum force is pointwisely first order consistent comparing to the scaled atomic total energy.
- The scaled continuum displacement field has first order modeling error wrt the atomistic $\| \cdot \|_\star$. 
Physical continuum boundary conditions

- In a 1D atomistic model, Stefanelli proved that the boundary needs to specify the length of the chain in direction $e_1$. \(^{17}\)

\[ y_{N,1} - y_{1,1} = \mu(N - 1)\varepsilon, \quad \mu \in (2/3, 1). \]

In direction $e_2$, simply apply the clamped condition

\[ y_{1,2} = D_1^\varepsilon y_{1,2} = y_{N,2} = D_1^\varepsilon y_{N,2} = 0. \]

This corresponds to suspending the atomistic sample. \(^{18}\)

- In the 2D continuum simulations, Zhang et al. 2014 obtained ripples with clamped boundary conditions applied to direction $e_3$. \(^{19}\)

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\(^{17}\) U. Stefanelli, et. al. arxiv.1802.05053, (2018)


Summary

- A continuum von-Kármán model is derived from atomistic descriptions by carefully specifying the scaling rules and homogenization.
- The continuum model is proved to be of second order in terms of energy, is of first order in terms of forces and displacement fields.
- A rigorous study of physical continuum boundary conditions is needed for both 1D and 2D atomistic and continuum models.
- Development of multiscale model is an ongoing project.
Thank you for your attention!

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